Infra-red microscopy study of laser-driven phase transformations in fused silica

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INTRODUCTION

Vitreous silica is the material of choice for transmissive and diffractive optics in high-power laser chains. The intensity delivered by high-power lasers for inertial confinement fusion (ICF) is limited by optical breakdown in the silica [1]. Laser-induced damage (LID) in fused silica windows is often observed to initiate at the rear surface. The LID threshold (LIDT) defines the fluence where permanent damage is observed at the surface of the fused silica window. Once damage is initiated, it propagates at sub-threshold fluence upon repetitive irradiation [2]. At higher fluence, damage initiates at the front surface. Damage at the rear surface is explosive and is characterized by extensive material cracking. At the front surface the laser beam drills a smooth-walled channel and redeposited material is observed around the drilled channel (Fig. 1) [2].

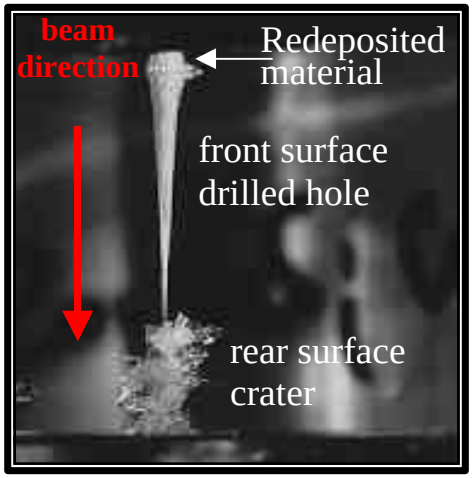

Fig. 1: Laser-damage morphologies.

The purpose of this study is to determine whether repetitive laser irradiation of fused silica cause structural changes in the amorphous network or transformations into high-pressure phases.

Experimental

Fused silica windows (Corning 7980) were irradiated with nanosecond pulsed lasers (λ=1064 and 355 nm). The rear-surface of the windows damaged both in the IR and the UV, while visible damage was observed on the front-surface only when the UV beam was used. The IR laser had a 4 mm x 3.5 mm square beam and 17-ns FWHM pulse duration. Under these conditions the LIDT of the rear-surface was approximately 55±5 J/cm^2. The UV laser had a Gaussian spatial profile (1/e^2 beam diamater=1.2 mm) and 8-ns FWHM pulse duration. The LIDT of the rear-surface under these conditions was 30±2 J/cm^2. The material analyzed for phase transformation was the white powder found inside the channel and at the bottom of the damage crater at the rear-surface. At the front surface the material analyzed for phase transformation was the recondensed material surrounding the drilled hole.

The samples were characterized using IR specular reflectance microscopy at Beamline 1.4.3 on the Advanced Light Source at Lawrence Berkeley National Laboratory. A thermal black body source was used to illuminate the samples. Combined with the microscope optics, the spatial resolution of the characterization is about 100 μm.

Results and Discussion

Fig. 2 is the reflectance spectrum of as-received polished silica surface and a rear-surface damage crater. The IR reflectance spectrum of the as-received surface is in agreement with the published spectrum of fused silica [3]. The main features are a peak centered around 790 cm^{-1} attributed to Si-O-Si rocking motion and a strong peak at 1123 cm^{-1} with a shoulder at 1236 cm^{-1} due to asymmetric stretching of tetrahedral Si-O units [3]. The silica inside the crater shows the emergence of a broad band at 900 cm^{-1}, whose intensity is comparable to the one of the Si-O stretch. The broad feature appearing at 900 cm^{-1} in rear-surface crater material corresponds to the vibration of non-bridging oxygen sites in the silica network, as shown by calculations of local vibrational modes [4] and spectroscopy studies of sol-gel silica [5]. The high pressure generated by the laser pulse as it is absorbed in the glass, causes Si-O bonds to break increasing the concentration of non-bridging oxygen atoms. As a result, the structure of silica is permanently densified.

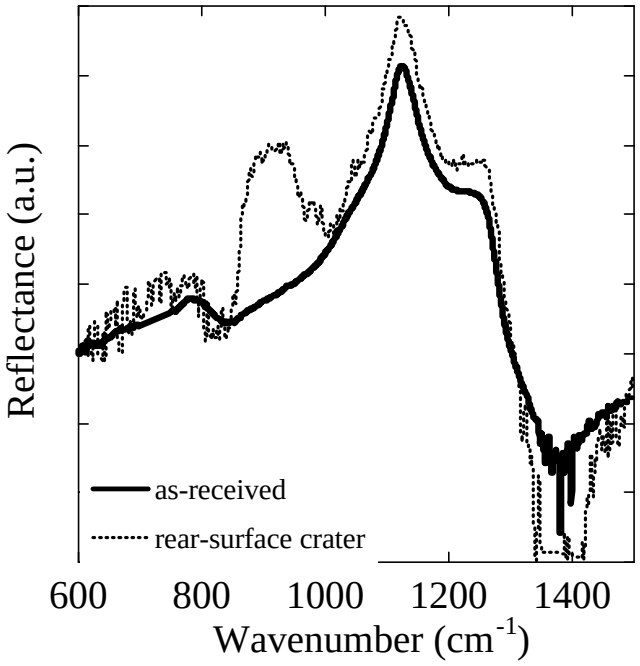

Fig.2: IR reflectance spectra of as-received and rear-surface laser-damage crater.

Fig. 3 is the spectrum of front surface redeposited material, as received silica and synthetic stishovite, a high-pressure polymorph of SiO2. In the front-surface redeposited material, the intensity of the 1236 cm^{-1} shoulder increases until it becomes a separate peak. This peak also narrows and shifts to higher wavenumbers reaching 1258 cm^{-1}. The intensity of the main 1123 cm^{-1} peak decreases until almost vanishing. The 1236 cm^{-1} shoulder is usually attributed to the LO vibration of the 1123 cm^{-1} asymmetric stretch peak. It has been shown that powder-like nature the material enhances the intensity of LO peaks [6]. At the same time, features in the 800 cm^{-1} 1050 cm^{-1} interval of the spectrum appear and gain strength. The most prominent feature is a strong and narrow peak that emerges at 1035 cm^{-1} and progressively shifts to 1020 cm^{-1}. Weaker peaks can be seen at 875 cm^{-1} and 665 cm^{-1}. The stronger peak at 1035 cm^{-1} is in good agreement with the LO component of the Eu vibration of stishovite. The weaker features correspond to other stishovite vibrations [7].

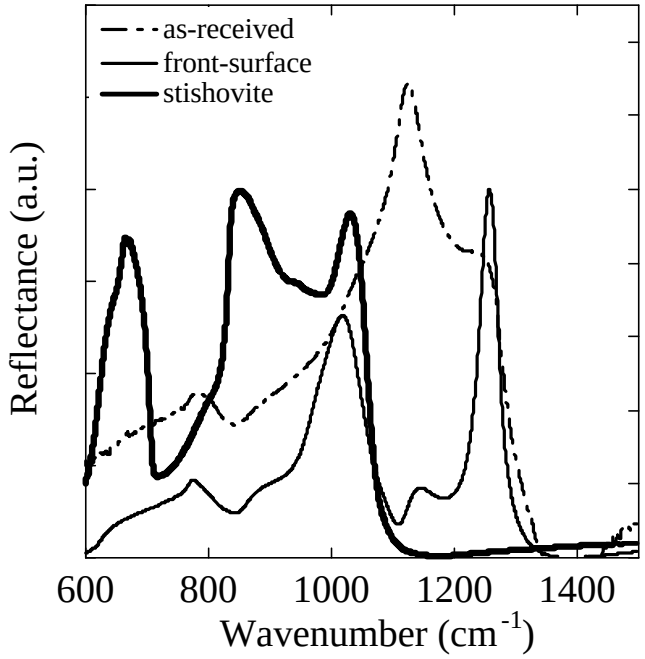

Fig. 3: IR reflectance spectra of as-received silica, front-surface redeposited material and synthetic stishovite.

Conclusions

Infra-red reflectance microscopy of laser-damaged silica shows that laser irradiation causes structural changes in the material. At the rear-surface, the pressure generated by the laser pulse breaks Si-O bonds generating non-bridging oxygen sites. At the front-surface, phase transformation of silica into stishovite is observed. This transformation is significant because stishovite is the only silica polymorph where Si is six-fold coordinated and has therefore very different electronic and optical properties than silica. Further investigation is needed to determine whether such a transformation can be detected in rear-surface damage material as well.

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